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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :  
JEAN-PIERRE CATINAT, ET AL. : EXAMINER: JOHN MABRY  
SERIAL NO: 10/534,502 :  
FILED: MAY 10, 2005 : GROUP ART UNIT: 1625  
FOR: PROCESS FOR THE :  
MANUFACTURE OF 1,2-EPOXY-3-  
CHLOROPROPANE

DECLARATION

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes **Jean-Pierre Catinat**, who deposes and states as follows:

1. That I am an inventor of the above-identified U.S. patent application, and that I am employed by Solvay (S.A.), Brussels.
2. That I am familiar with the English language.
3. That the above-identified U.S. Patent Application describes and claims a process for the manufacture of 1,2-epoxy-3-chloropropane comprising reacting allyl chloride and hydrogen peroxide in an epoxidation medium comprising at least one liquid phase and in the presence of a catalyst comprising a zeolite, wherein the pH of the liquid phase is controlled and maintained at a value of greater than or equal to 1.5 and less than 4.8.
4. It is my understanding that the U.S. Examiner has taken the position that it would be routine experimentation for an artisan of ordinary skill to modify pH in order to determine optimum range. It is my further understanding that the U.S. Examiner has taken the position

that a combination of the teachings in Strebel (U.S. patent 6,288,248), Nakanishi (Japan 04327582) and Gilbeau (U.S. patent 6,063,941) would have lead one of ordinary skill in this art to expect what I and my coinventor have found - that a process for the manufacture of 1,2-epoxy-3-chloropropane comprising reacting allyl chloride and hydrogen peroxide in an epoxidation medium comprising at least one liquid phase and in the presence of a catalyst comprising a zeolite, wherein the pH of the liquid phase is controlled and maintained at a value of greater than or equal to 1.5 and less than 4.8, provides a higher selectivity without excessively reducing the activity of the catalyst in comparison with a process carried out without control of the pH.

5. That I have reviewed Strebel, Nakanishi, and Gilbeau and that there is nothing in any of these references, or their combination, that would lead one of ordinary skill in the art to expect the higher selectivity obtained with the present invention accomplished without excessively reducing the activity of the catalyst.

As explained in the specification, we have found that when the pH is controlled and maintained at a value of greater than or equal to 1.5 and less than 4.8, the selectivity of the epoxidation is higher without excessively reducing the activity of the catalyst, in comparison with a process carried out without control of the pH. This is demonstrated by the Examples and comparative Examples at specification pages 6-9 and is summarized in Tables 1-3, which demonstrate the greatly improved selectivity (Tables 1 and 2) of the invention as well as improved catalyst life (Table 2; time) when pH is controlled according to the invention as compared to the case where pH is not regulated:

TABLE 1

Time (h)		Example 1 With regulation	Example 2 Without regulation
25	Conversion (%)	95.3	96.9
	EPI/C3f selectivity (%)	96.9	93.7
50	Conversion (%)	89.3	92.1
	EPI/C3f selectivity (%)	97.2	94.3
78	Conversion (%)	83.3	85.1
	EPI/C3f selectivity (%)	97.4	94.4

TABLE 2

	Example 3 T°: 65° C. With pH regulation	Example 4 T°: 55° C. Without pH regulation
Degree of conversion H <sub>2</sub> O <sub>2</sub> (%)	92.0	92.2
EPI/C3f selectivity (%)	96.3	94.4
Time (h)	195	53

Strebel and Nakanishi are silent with regard to pH control/effect, and Gilbeau does not relate to the effect of pH during a chemical reaction. Even if it were accepted that it is obvious to vary pH within a given range, none of Strebel, Nakanishi, and Gilbeau would have predicted that when the pH of the liquid phase is controlled and maintained at a value of greater than or equal to 1.5 and less than 4.8 as claimed in this patent application that a double benefit would be obtained in the form of both improved selectivity and improved catalyst life.

6. During prosecution in Europe WO 99/48882 (D1), EP-A-1 072 600 (D2), and EP-A-1 085 017 (D3) were cited against the corresponding European application. It is my understanding that these references and their list of equivalents have been filed in this U.S. patent application.

References D1 to D3 are important because all their examples teach that a decrease in the pH of the reaction medium to a value included in the range claimed according to this invention has the effect of substantially decreasing the selectivity of the epoxidation reaction. In addition, none of the examples of these three documents makes it possible to achieve, at the same time, a good selectivity of the epoxidation reaction without decreasing the speed/rate of conversion of the hydrogen peroxide.

In D1, a decrease in pH from 5.5 to 4.0 has the effect of decreasing the selectivity from 90.7 to 84.0% (Examples 1 and 2). In D2, a pH of 6.0 for the reaction medium leads to a selectivity of 97.2% and a rate of conversion of hydrogen peroxide of 96% after 30 hours (Example 3), whereas, if the pH of the reaction medium is decreased to 4.5, these percentages become respectively 80% and 75% (Example 2). In D3, a pH of 5.46 for the reaction medium leads to a selectivity of 78.4% (Example 12), decreasing to 66.8% when the pH of the reaction medium is decreased to 4.39 (Example 7), the rate of conversion of the hydrogen peroxide being unchanged (58% after approximately 5 hours).

7. These references thus show that what is obtained with the present invention - high selectivity of the epoxidation reaction without decreasing, or even while increasing, the rate of conversion of the hydrogen peroxide - is not expected and, in fact, is contrary to what is expected from the references. For example, comparison of examples 1 and 3 (with adjustment of the pH) with examples 2 and 4 (without adjustment of the pH) (See Tables 1 and 2 above) clearly shows the positive effect of pH adjustment on the selectivity of the epoxidation reaction, while the rate of conversion of the hydrogen peroxide is only very slightly reduced (from 85.1 to 83.3% after 78 hours in examples 1 and 2) or is even improved (rate of conversion of 92% after 195 hours with adjustment of the pH in comparison with the same rate of conversion but after only 53 hours without pH adjustment). These unexpected results are very important in this art, as catalyst life and selectivity are important factors in the

economic viability of a process, especially a process for an intermediate commodity such as epichlorohydrin.

8. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

9. Further deponent saith not.

Catinat Jean-Pierre

JEAN-PIERRE CATINAT

DATE

July 4, 2008